

Deposition of biaxially aligned YSZ layers by dual unbalanced magnetron sputtering.

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Abstract. Biaxially aligned YSZ (Yttria Stabilised Zirconia) layers were deposited by unbalanced magnetron sputtering, in a dual magnetron geometry. The unbalanced magnetrons were mounted in such a way that the angle between the target- and substrate normal was 55° for both magnetrons. The target-substrate distance was 13 cm for both magnetrons. A better homogeneity in deposition rate and biaxial alignment was obtained with respect to depositions with one unbalanced magnetron. The YSZ layers were characterized by XRD $\theta/2\theta$ and (111) pole figures and showed a [001] out-of-plane orientation and a [110] in-plane orientation. The best biaxially aligned YSZ layers obtained so far, showed a FWHM of 21° in (111) pole figures. The influence of the magnet configuration (closed-field or mirror-field) and sputter conditions on the biaxial alignment was investigated. Gauss and Langmuir probe measurements were performed to investigate the influence of the magnet configuration and sputter conditions on the plasma density and the magnetic field lines.

1. Introduction

Biaxially aligned layers have a preferential out-of-plane (perpendicular to the substrate) orientation, as well as a preferential in-plane (parallel to the substrate) orientation. These biaxially aligned layers have specific advantages, especially in applications where elimination of high angle grain boundaries improves product properties. One of the potential applications is a buffer layer for coated conductors, because the superconducting properties of a $\text{REBa}_2\text{Cu}_3\text{O}_{7-x}$ ($\text{RE} = \text{Y}, \text{Nb}, \dots$) layer strongly improve when high angle grain boundaries are minimized [1].

To obtain biaxially aligned layers on a polycrystalline or amorphous substrate, special deposition techniques such as ion beam assisted deposition (IBAD) [2] and inclined substrate deposition (ISD) [3] are necessary. In our group, an unbalanced magnetron type II (in the classification of Windows and Savvides [4], more specific when the outer magnetic field is stronger than the inner magnetic field) is used to deposit biaxially aligned Yttria Stabilized Zirconia (YSZ) layers [5]. Up to now, a single sputter magnetron was used [6], but in this research a dual magnetron geometry is investigated in order to obtain a better homogeneity in the deposited layers.

2. Deposition with a single magnetron.

The sputter source consists of a type II unbalanced magnetron. The substrate (mirror like polished stainless steel) was mounted on a rotatable substrate holder, which could be tilted over an angle α (α is the angle between the substrate normal and the direction of the incoming material flux). The target-substrate distance was 13 cm. The 2 inch target (Zr/Y 55/45 Wt%) was sputtered in reactive DC mode, with a mixture of Ar and O₂. The sputter power was kept at 250 Watt during deposition. The base pressure before deposition was below 10^{-3} Pa and the sputter pressure was 0.25 Pa. These sputter conditions resulted in a deposition rate of ~ 45 nm/min.

The optimal angle α was investigated by varying the tilting angle of the substrate. The deposition time was chosen in order to obtain layers of about 1.15 μm thick. These layers were characterised by $\theta/2\theta$ and by (111) pole figures to investigate respectively the out-of-plane and in-plane alignment. The results of these XRD measurements can be seen in table 1. A (111) pole figure of the layer deposited at $\alpha = 55^\circ$, together with the direction of incoming material flux is shown in figure 1.

The layer deposited on a non-tilted substrate ($\alpha = 0^\circ$) shows a nearly random out-of-plane alignment (especially [002] and [311] but also small peaks of [111] and [220]). No in-plane orientation was measured for this layer.

All YSZ layers deposited on a tilted substrate exhibit a [002] out-of-plane alignment and a [110] in-plane alignment (in the definition of Sonnenberg et al. [7]). In table 1, it is clear that the in-plane alignment improves, by increasing the substrate tilting angle α . It is believed that the in-plane alignment is partially caused by shadowing. This shadowing process will be more efficient when the angle α is increased.

Table 1. XRD results of the depositions with one magnetron on varying angle.

Angle α	Out-of-plane orientation	In-plane alignment FWHM of (111) pole
0°	nearly random	/
30°	[002]	30.3°
45°	[002]	22.8°
50°	[002]	21.9°
55°	[002]	20.3°

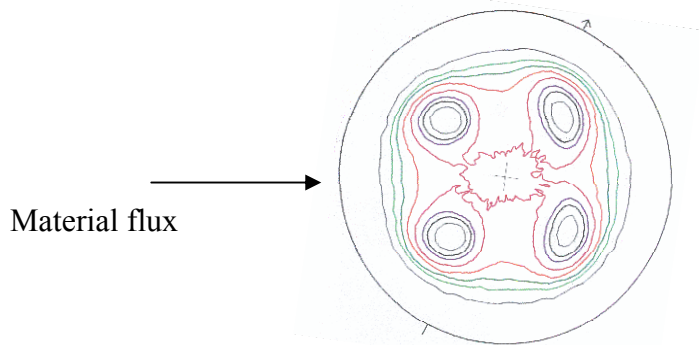


Figure 1. (111) pole figure of an YSZ layer deposited with one magnetron, $\alpha = 55^\circ$.

A major problem of depositing layers on a tilted substrate with one single deposition source is the inherent inhomogeneity of the layer thickness and in-plane alignment. This is illustrated by figure 2.

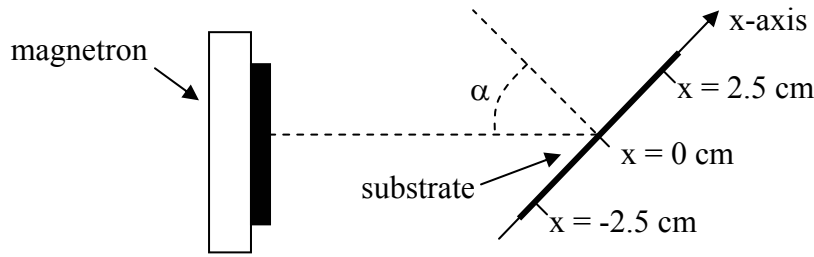


Figure 2. Deposition geometry with a single magnetron.

All results of table 1 are for depositions on a substrate at position $x=0$ (see figure 2). But when depositing on a larger substrate, there will be an inhomogeneity in thickness and in-plane alignment over the length of the substrate (different x -position) .

The layer thickness is more or less proportional to $1/R^2$ with R the distance between the substrate and the centre of the target, so the YSZ layer thickness will vary with the position x . Also a large difference in the in-plane alignment with varying x -position is observed, as shown in table 2.

Table 2. XRD results for deposition with one magnetron at different position x .

Angle α	x -position	Out-of-plane orientation	In-plane alignment FWHM of (111) pole
55°	-2.5 cm	[002]	18.5°
55°	0 cm	[002]	20.3°
55°	2.5 cm	[002]	34.3°

This inherent inhomogeneity in layer thickness and in-plane alignment makes scaling-up of the deposition process with one source difficult. A dual deposition geometry was used to avoid these inhomogeneities, allowing to deposit homogenous layers on a larger area. This deposition geometry is described in section 3.

3. Deposition with a dual magnetron geometry

3.1. Deposition geometry

The best in-plane alignment, obtained by using a single magnetron was obtained by doing deposition on a substrate with tilting angle $\alpha = 55^\circ$, as can be seen in table 1. So, if we want to use two magnetrons at the same time, we will have to mount them in such a way that the angle between the target- and substrate normal is 55° for both of them. In order to be able to compare the depositions with one magnetron and the depositions with the dual geometry, the magnetrons in the dual geometry were fixed in such a way that the target-substrate distance is 13 cm for both of them.

The sputter power was 250 Watt for each magnetron, and the sputter pressure was varied between 0.2 and 0.3 Pa. The used substrate is now glass.

When using two magnetrons, two different magnet configurations are possible. The directions of the magnetic field lines caused by these two magnet configurations were measured by a Gauss probe (Bell 640 Incremental). The two magnet configurations and the direction of the measured magnetic field lines are shown in figures 3 and 4.

In the mirror-field configuration, the magnets of both circular magnetrons were mounted in the same way (two magnetrons with the same magnetic polarity). On the other hand, the magnets of both magnetrons in closed-field configuration were mounted reversed.

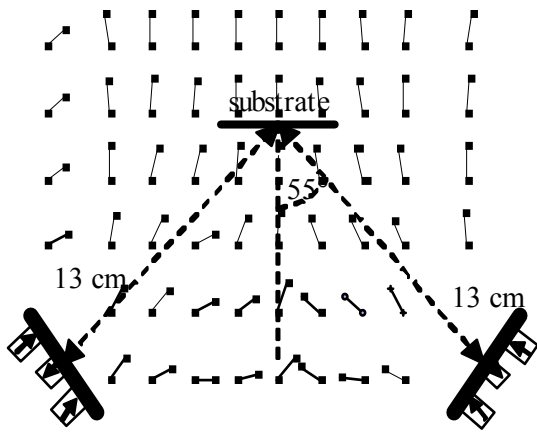


Figure 3. Magnetic field lines of the mirror-field configuration.

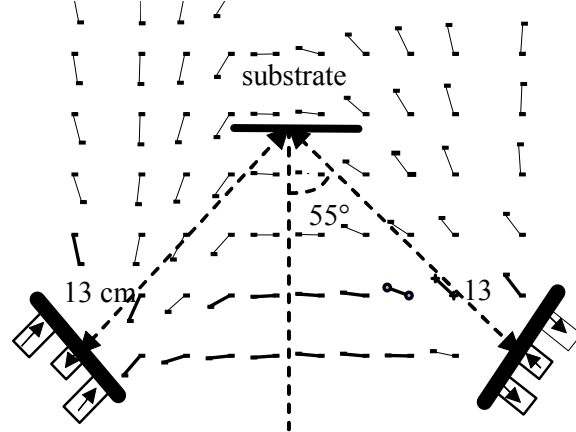


Figure 4. Magnetic field lines of the closed-field configuration.

In the closed-field system the field lines close in across the chamber (see figure 4). Electrons following these field lines are available to undergo ionizing collisions and, thus, maintain a high plasma density in the vicinity of the substrate. In the mirror-field, the electrons are directed towards the chamber walls (see figure 3), resulting in a lower plasma density

3.2. Plasma characteristics

The plasma characteristics in the vicinity of the substrate were determined by Langmuir probe measurements. The electron temperature T_e , plasma potential V_p , floating potential V_f and ion density n_i were determined by a small cylindrical Langmuirprobe (Hidden analytical).

The $V_p - V_f$ remained 13 Volt during all deposition conditions with the dual magnetron system. Also no influence of the variation in pressure (0.2 or 0.3 Pa) and magnet configuration on the T_e was seen. On the other hand, a clear influence of the magnet configuration on the ion density was seen.

The ion density in the vicinity of the substrate seemed to be almost two times higher under the closed-field configuration as compared with the mirror-field configuration, which is in agreement with literature [8].

3.3. Deposition results

The use of two magnetrons at the same time seemed to have no influence on the out-of-plane alignment because all the YSZ layers had preferential [002] out-of-plane orientation. All layers deposited by the dual geometry showed a good [110] in-plane alignment as proved by (111) pole figures. The results of the (111) pole figures of the sample at x-position - 0.5 cm are summarized in table 3. A (111) pole figure of the layer deposited 0.3 Pa and mirror-field is shown in figure 5.

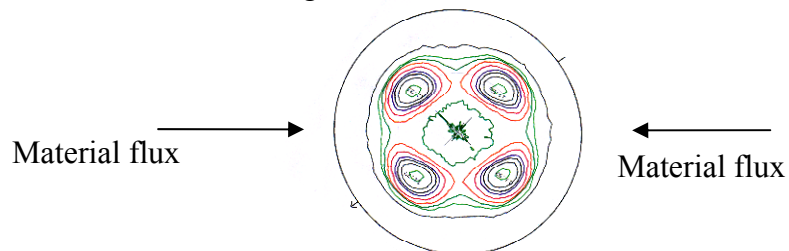


Figure 5. (111) pole figure of a YSZ layer deposited with two magnetrons.

Table 3. XRD results of the depositions with the dual geometry system.

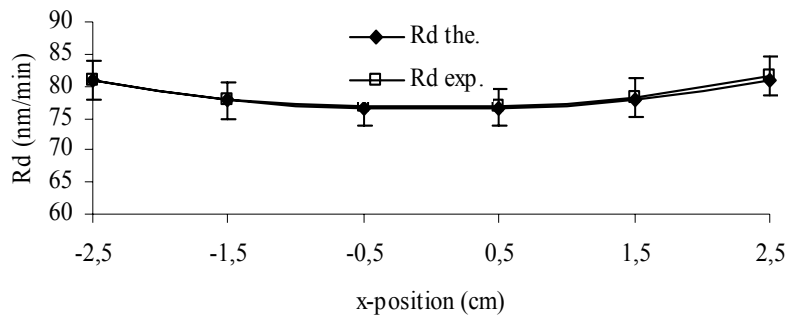
Pressure Pa	Magnet configuration	Out-of-plane orientation	In-plane alignment FWHM of (111) pole
0.2	mirror	[002]	25.3°
0.2	closed	[002]	21.9°
0.3	mirror	[002]	25.2°
0.3	closed	[002]	22.6°

No influence of the sputter pressure on the in-plane alignment can be seen in table 3. However, a strong influence of the magnet configuration on the in-plane alignment is visible. The closed-field magnet configuration seems to yield a better in-plane alignment. It is believed that this better in-plane alignment is caused by the higher plasma density in the vicinity of the substrate during deposition in the closed-field configuration. A higher plasma density can cause a higher surface mobility of the adatoms on the growing YSZ layer, which can benefit the biaxial alignment mechanism.

The goal of the development of this dual magnetron system was to improve the layer homogeneity. As mentioned in section 2, the deposition speed (R_d) is proportional to $1/R^2$ with R the substrate-target distance. However, when depositing layers with two magnetrons, we have to add up the deposition speeds of the two separate magnetrons. The results of the calculation of the deposition speeds ($R_d \sim 1/R_1^2 + 1/R_2^2$, with R_1 and R_2 respectively the distance to target 1 and target 2) at different x-positions of the substrate, and the actually measured deposition speeds are shown in figure 6. The measured deposition speeds are of the YSZ layer deposited by 0.3 Pa and the mirror-field configuration.

As can be seen in figure 6, the experimental deposition speed fits well with the theoretical $1/R^2$ dependence, so the YSZ layer thickness deposited with two sources is much more homogeneous in comparison to layers deposited with one source.

An improvement of the homogeneity of the in-plane alignment was also established, as can be seen in figure 7. The YSZ layer on a substrate longer than 5 cm, deposited by two 2" magnetrons varied only by 2° FWHM in in-plane alignment.

**Figure 6.** Comparison of the theoretical and experimental deposition speed.

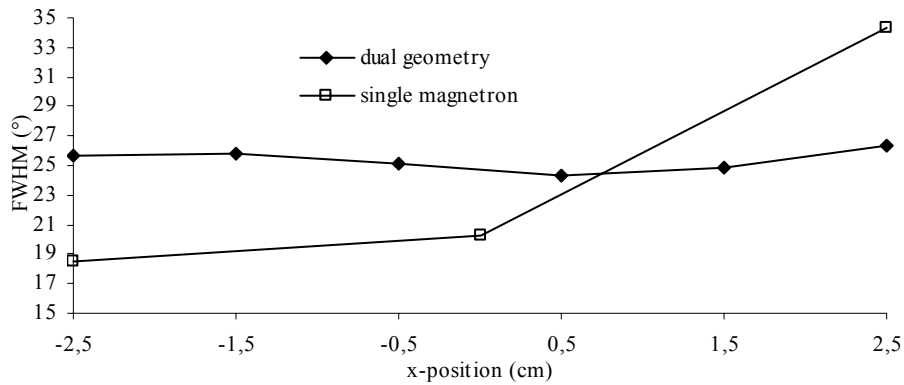


Figure 7. Comparison of the in-plane alignment at several x-positions for layers deposited with one single magnetron and layers deposited with the dual geometry.

4. Conclusions

Biaxially aligned YSZ layers were deposited by dc unbalanced magnetron sputtering. The influence of the target-substrate angle α was investigated by deposition with one single magnetron. It seemed that the in-plane alignment improves, by increasing the target-substrate angle, which is probably due to a more pronounced shadowing effect.

A dual magnetron system was explored to improve the layer homogeneity. The biaxial in-plane alignment of YSZ layers deposited with two magnetrons is a little less good than the in-plane alignment of layers deposited with one source. However, a very good homogeneity in layers thickness and in-plane alignment was obtained.

Further optimisation of the in-plane alignment of YSZ layers deposited by unbalanced magnetron sputtering is needed.

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